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FIRST RESULTS OF REACTION PROPAGATION RATES IN HMX AT HIGH PRESSURE

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Abstract. We have measured the reaction propagation rate (RPR) in weapons-grade, ultrafine octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX) powder in a diamond anvil cell over the pressure range 0.7-35 GPa. In order to have a cross-comparison of our experiments, we carried out a series of experiments on nitromethane (NM) up to 15 GPa. Our results on NM are indistinguishable from previous measurements of Rice and Folz. In comparison to high-pressure NM, the burn process for solid HMX is between 5-10 times faster at pressures above 10 GPa.

INTRODUCTION

There is a strong interest in first-principles modeling of chemical reactions in high explosive (HE) materials. However, the validation of these models requires experimental data at the appropriate pressure and temperature conditions of the reactions of interest. Because these reactions occur over time scales of microseconds, they have to a great degree resisted experimental characterization of the fundamental processes governing combustion and detonation. The diamond anvil cell (DAC) is well suited for studying these reactions because it provides a high-pressure, variable-temperature sample environment, as well as an optically clear window for spectroscopic study of reactions within the DAC. The reaction propagation rate (RPR) of an HE material can be studied directly by confining the material within the DAC and initiating combustion with a focused laser pulse. Our experimental approach is a modification of the earlier work of Rice and Foltz (1). Here we report the first results of the RPR measurements on octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX) over the pressure range 0.7-35 GPa.

EXPERIMENTAL

The experimental setup is presented in Figure 1. The apparatus and procedure employed were similar to those used by Foltz in RPR measurements of the high explosive 1,3,5-triamino-2,4,6-trinitrobenzene (TATB), except the measurements described here were done employing transmitted light rather than reflected light (2). Samples were contained in a diamond anvil cell (DAC) consisting of two counter-opposed 0.25 carat diamonds with culet diameters of 0.5-1.0 mm. Lateral confinement of the 100-150 μm thick samples was achieved using Inconel or rhenium gaskets with 150-400 μm hole diameters. Ruby powder was deposited onto the surface of the diamond nearest the streak camera for determination of the initial pressure using the ruby fluorescence pressure scale. Samples consisted of weapons-grade, ultrafine HMX (lot B-881-wet) containing less than 7% RDX, with a grain size of ~3 μm. In addition, RPR studies of nitromethane (NM) were conducted for comparison with the previous data of Rice and Foltz (1).

Both sample illumination and excitation for ruby fluorescence were provided by an argon ion laser operating at 488 nm (Lexel model 95). The cw 488 nm beam was passed through a beam expander and focused into the DAC in order to fully illuminate the sample. Sample ignition was provided by a Q-switched Nd:YAG laser (New Wave MiliLase II-20) frequency-doubled to 532 nm, with pulses of ~9 ns duration. During optical alignment, the 532 nm pulse energies were kept below 0.1 μJ to prevent accidental ignition. The 532 nm beam was made collinear with the 488 nm beam using a quartz beamsplitter, and was focused to a ~5 μm spot size in the center of the sample region illuminated by the 488 nm beam. Transmitted light from the sample was magnified (~10x) and focused onto a 10-50 μm-wide slit, and was then magnified (9.5x) after the slit. For ruby fluorescence measurements, the emission was focused onto the entrance slit of a Kaiser Optics f/1.8 spectrograph, and detected by a liquid-nitrogen cooled CCD camera (Princeton Instruments).

After pressure measurements, the laser speckle pattern from the DAC provided by the 488 nm beam was directed to an EG&G L-CA-20 electronic streak camera (Polaroid film type 57, 3000 speed) operating at streak rates between 1.8 and 10 μs. A glass slide in the 532 nm beam was used to direct a portion of the ignition pulse to a photodiode in order to synchronize the pulse with the streak camera. Ignition pulse energies were determined by a Molectron EPM 2000 energy meter, and were in the range of 1-10 μJ. A holographic notch filter placed before the streak camera slit was used to filter the 532 nm light to prevent over-exposure of the streak image. Typical streak images are shown in Figure 2.

RESULTS AND DISCUSSION

The experiments on NM were performed in order to compare our RPR data with known values. Figure 3 presents a comparison of burn rates obtained by Rice and Foltz (1) with those obtained in our laboratory. The figure demonstrates that our results are indistinguishable to those previously obtained, suggesting that our experimental method is robust for the study of HE materials.

Table 1 presents the RPR values obtained in the present study on HMX in the pressure range 0.7-35 GPa. These data indicate that the burn rate of HMX is between 5 and 10 times that of NM at pressures above 10 GPa. Furthermore, the data also demonstrate that at HMX burns over an order of magnitude faster than TATB in this pressure regime (2).

TABLE 1. Reaction Propagation Rates for HMX

Pressure (GPa)	Rate (m/s)
0.7	4.3 ± 0.8
1.7	9.5 ± 2.5
2.8	11.0 ± 2.0
3.7	9.3 ± 0.9
7.7	53.0 ± 5.3
9.1	152 ± 5.5
11.0	186 ± 19
13.0	228 ± 18
14.0	242 ± 23
21.0	222 ± 23
25.0	257 ± 26
35.0	641 ± 70

The dependence of the RPR of HMX on pressure is presented in Figure 4. Also presented are the calculated burn rates for HMX obtained from Reaugh. While the calculation consistently underestimates the observed burn rate, the two plots have similar slopes, indicating that the calculation follows the observed progression of burn rate with pressure.

REFERENCES

1. Rice, S.F, and Foltz, M. F., *Combustion and Flame* **87**, 109-122 (1991).
2. Foltz, M. F., *Propellants, Explosives, Pyrotechnics* **18**, 210-216 (1993).
3. Reaugh, J. E., unpublished results.

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Farber et al, figure captions

Figure 1 caption:

Schematic layout of RPR experimental apparatus. The argon ion laser provides sample illumination and excitation for ruby fluorescence pressure measurements, while the sample ignition pulse is provided by the Nd:YAG laser. The argon ion laser speckle pattern is imaged onto the streak camera slit, and disturbances to the speckle pattern caused by reaction of the sample are recorded.

Figure 2 caption:

2a. Streak record of a reacting HMX sample at 21 GPa. The vertical dimension is time, where the streak rate is 3.2 μs, and the horizontal dimension is distance. The parallel vertical lines are due to the undisturbed laser speckle pattern being streaked in time. Deflagration within the sample disturbs the speckle pattern from the point where the ignition pulse strikes the sample. The disturbance moves outward from this point, resulting in the pattern shown here. The corresponding reaction propagation rate is 223 m/s.

2b. Streak record of a reacting HMX sample at 35 GPa, with a streak rate of 3.2 μs (for direct comparison with fig. 2a). The corresponding reaction propagation rate is 641 m/s.

Figure 3 caption:

Pressure dependence of the reaction propagation rate for nitromethane. Points with error bars were obtained by Rice and Foltz (1991), while the data indicated by the circles were obtained in our laboratory.

Figure 4 caption:

Pressure dependence of the reaction propagation rate for HMX. Experimental data obtained in our laboratory are indicated by the circles, while the squares indicate calculated rates.

Farber et. al. Figure 1

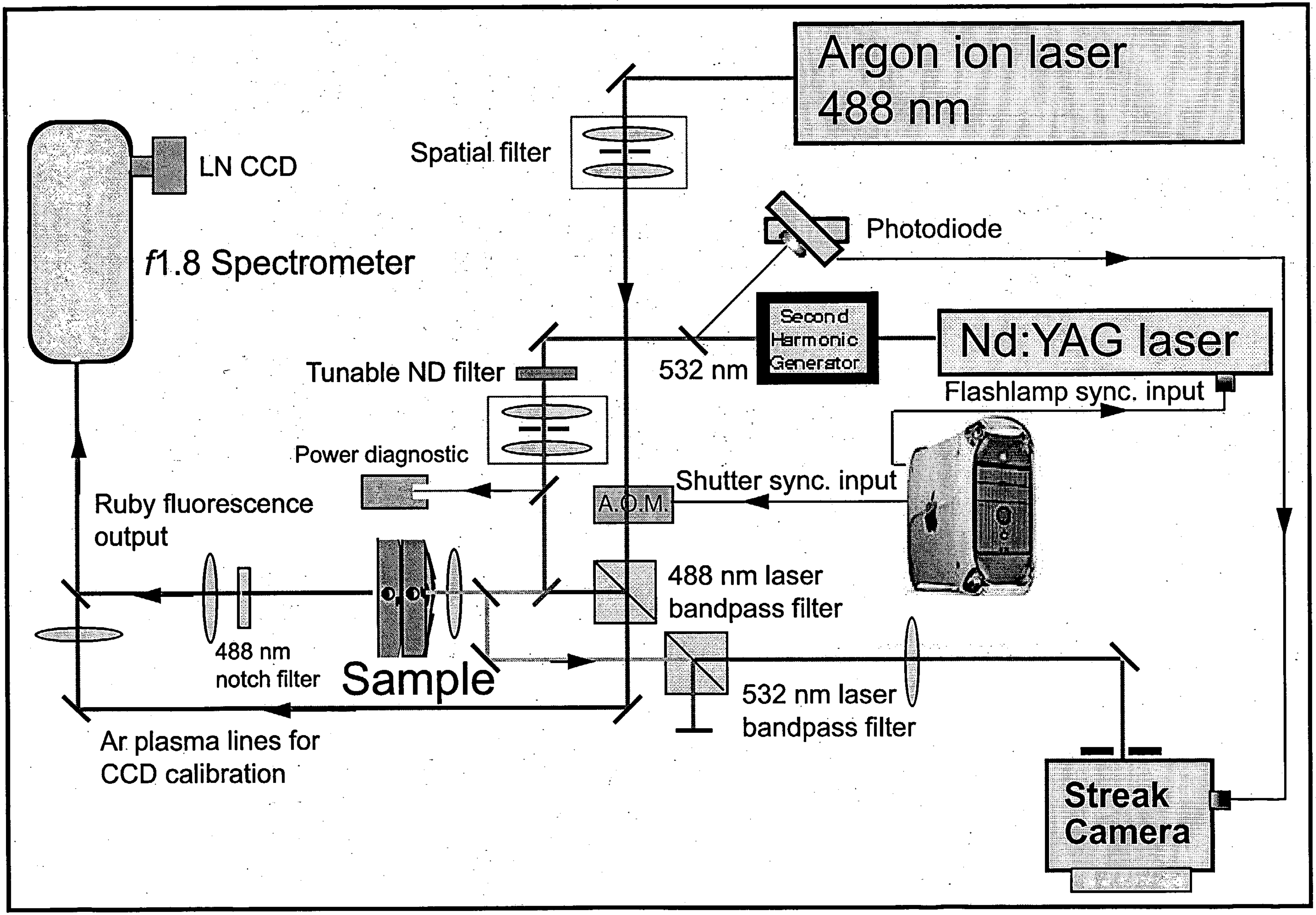

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Figure 2a

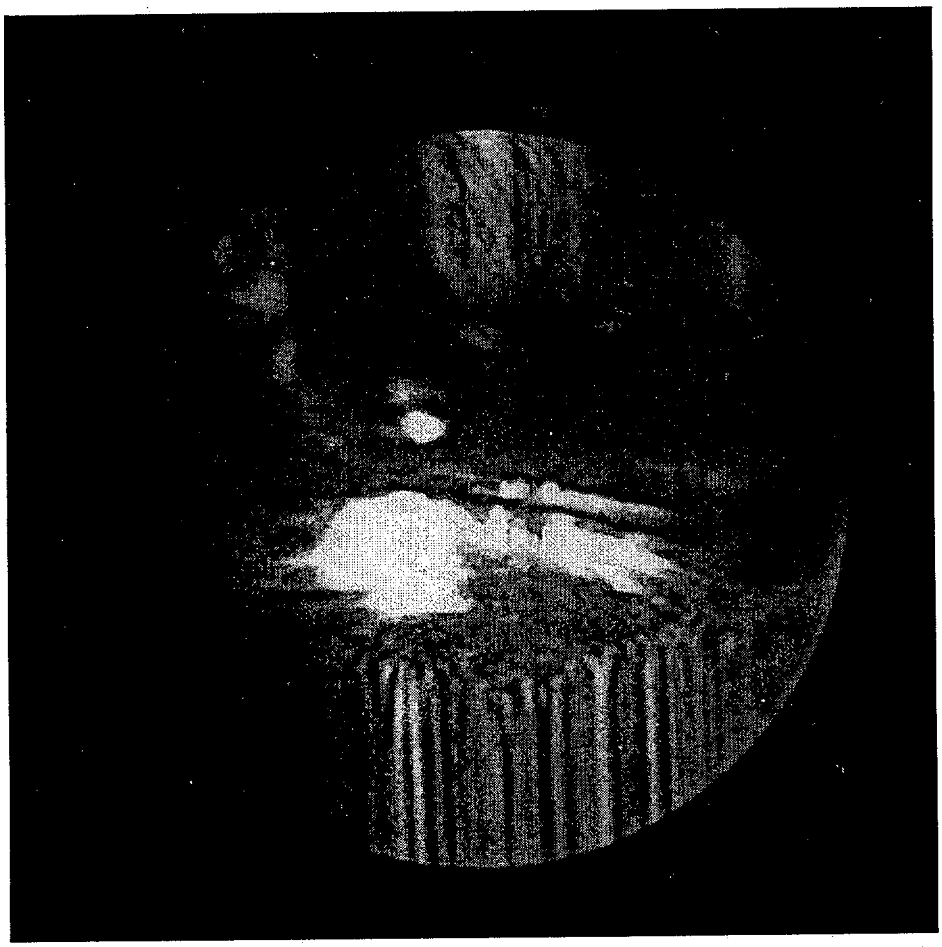

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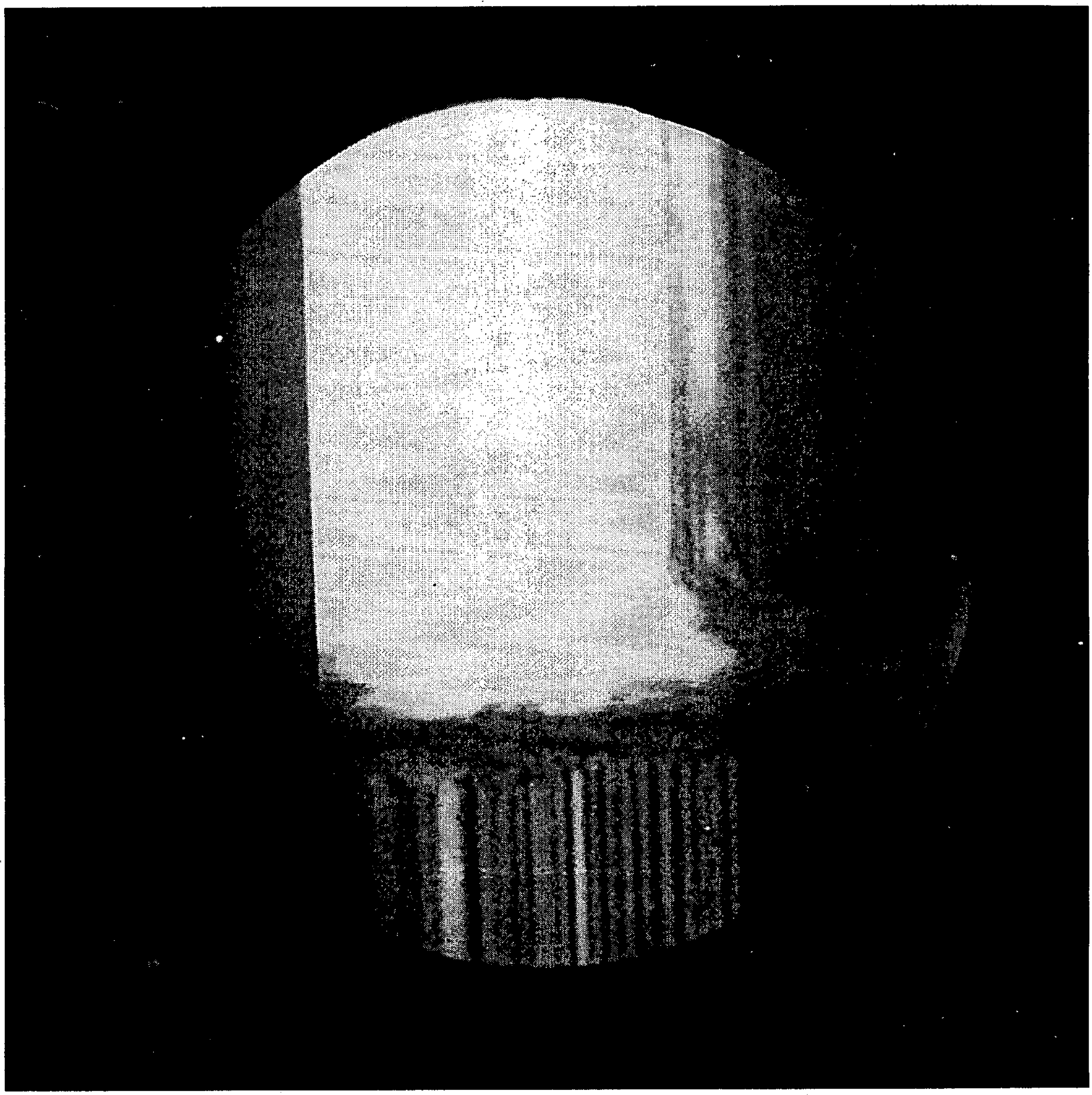
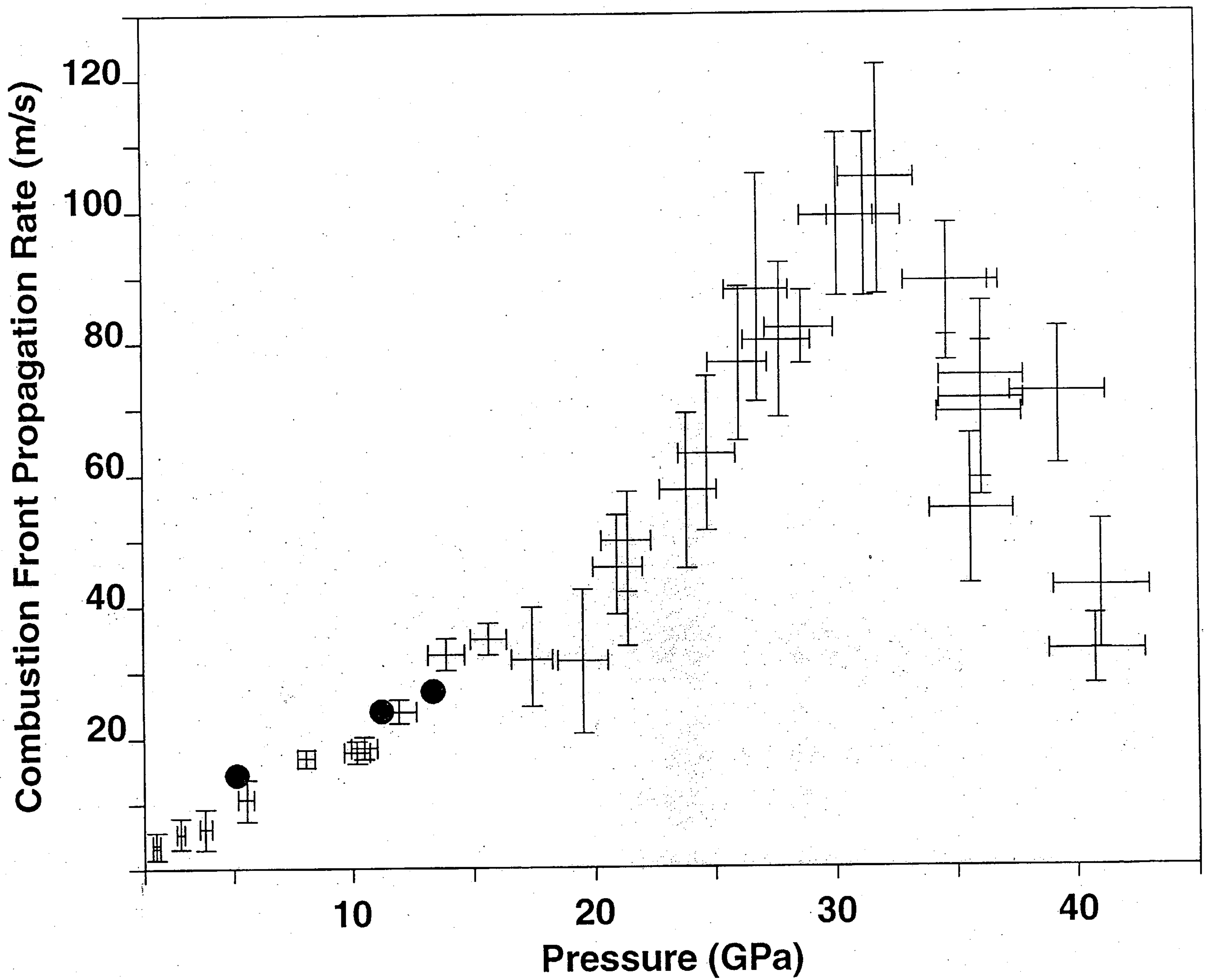
Figure 2b



Farber et.al. Figure 3



Tarber et. al. Figure 4